

## IGNITION BEHAVIOR OF PULVERIZED COALS

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### INTRODUCTION

We present data from a laser-based experiment used to measure the ignitability of pulverized coals in a room-temperature gas environment. The absence of hot furnace walls surrounding the test section allowed for optical detection of the ignition process. The experimental parameters studied include the coal type, oxygen concentration and particle size. The results show clearly that ignition reactivity is strongly dependent on coal type, and that the ignition rate constants determined are consistent with published data for overall combustion reactivity. The data also show convincingly that particle-to-particle variations in physical and/or chemical property of the fuel must be accounted for in order to model the ignition data correctly, and to accurately describe their ignition reactivity. We present a distributed activation energy model which accomplishes this goal.

### EXPERIMENT

The experiment is similar to one described in detail elsewhere,<sup>1</sup> so only a brief description is given here. Figure 1 presents a schematic of the laser ignition experiment; the inset shows the details around the test section. Sieve-sized particles were dropped through a tube into a laminar, upward-flow wind tunnel with a quartz test section (5 cm square cross-section). The gas was not preheated. The gas flow rate was set so that the particles emerged from the feeder tube, fell approximately 5 cm, then turned and traveled upward out of the tunnel. This ensured that the particles were moving slowly downward at the ignition point, chosen to be 2 cm below the feeder-tube exit. A single pulse from a Nd:YAG laser was focused through the test section, then defocused after exiting the test section, and two addition prisms folded the beam back through the ignition point. Heating the particles from two sides in this manner achieved more spatial uniformity and allowed for higher energy input than a single laser pass. For nearly every case, two to five particles were contained in the volume formed by the two intersecting beams, as determined by previous observation with high-speed video.<sup>2</sup>

The laser operated at 10 Hz and emitted a nearly collimated beam (6 mm diameter) in the near-infrared (1.06  $\mu\text{m}$  wavelength). The laser pulse duration was  $\sim 100 \mu\text{s}$  and the pulse energy was fixed at 830 mJ per pulse, with pulse-to-pulse energy fluctuations of less than 3%. The laser pulse energy delivered to the test section was varied by a polarizer placed outside of the laser head; variation from 150 to 750 mJ was achieved by rotating the polarizer. Increases in the laser pulse energy result in heating of the coal particles to higher temperatures. At the ignition point the beam diameter normal to its propagation direction was  $\sim 3 \text{ mm}$  on each pass of the beam. An air-piston-driven laser gate (see Fig. 1) permitted the passage of a single pulse to the test section. The system allowed for control of the delay time between the firing of feeder and the passage of the laser pulse. Finally, ignition or nonignition was determined by examining the signal generated by a high-speed silicon photodiode connected to a digital oscilloscope, as described elsewhere.<sup>1</sup>

We report here the ignition behavior of two coals: one medium-volatile bituminous, and one high-volatile bituminous. Both samples were obtained from the Penn State University Coal Sample Bank, and the reported proximate and ultimate analyses are shown in Table 1. The coals were sieve-sized using a Ro-Tap shaker to -120/+140 mesh (106-125  $\mu\text{m}$  diameter), and dried at 70°C under vacuum for at least 12 hours prior to each day's experiment.

### RESULTS

Each day's experiment was conducted as follows: After choosing the coal and oxygen concentration to examine, the coal was loaded into the batch-wise feeder. The delay time between the triggering of the feeder and the appearance of the coal batch at the feeder tube exit was measured by visual observation in conjunction with a stop watch; typical values were  $\sim 2.9 \text{ s}$ . The delay time was then programmed into the device which triggered the laser gate. The gas flow rate needed to achieve a drop distance of  $\sim 5 \text{ cm}$  for the coal batch was also determined by visual observation. Finally, a laser pulse energy was chosen, and the experiment commenced. At each set of operating conditions (coal type and size, oxygen concentration, and laser energy), 20 attempts at ignition were made in order to measure the

ignition frequency, or probability, which is the parameter sought from these studies. Mapping this ignition frequency over a range of laser pulse energy produces an ignition-frequency distribution.

Such a frequency distribution is shown in Fig. 2 for the Pittsburgh #8 coal. It can be seen that at each oxygen concentration, ignition frequency increases monotonically over a range of increasing laser pulse energy. Below this range the ignition frequency is zero, and higher energies result in 100% ignition frequency. This behavior is due to the fact that, within any coal sample, there exists a variation of reactivity among the particles.<sup>3</sup> Thus, in this experiment, in which a batch of perhaps several hundred particles of a sample is dropped into the test section but only a few are heated by the laser pulse, there is an increasing probability (or frequency) as the laser energy is increased that at least one of the heated particles is reactive enough to ignite under the given conditions.

The repeated distributions under 100% oxygen, measured on separate days, show the excellent repeatability of this experiment; the most important factor for reproducibility is the moisture content of the sample.

Figure 2 also shows the effect of oxygen concentration: As oxygen level is decreased from 100% to 75%, and then to 50%, the frequency distribution shifts to higher laser energies or, equivalently, higher particles temperatures, as expected. This is consistent with ignition theory since at decreased oxygen levels, higher temperatures are necessary to achieve the equality between heat generation by the particles (due to chemical reactions) and heat loss from the particles. This equality is the minimum requirement for ignition, and is termed 'critical ignition.' The shift in distribution can be viewed in two ways: First, for a fixed laser pulse energy, a decrease in oxygen level leads to a decrease in the ignition frequency, all else being the same; second, a decrease in oxygen implies that a higher laser pulse energy is needed, in order to achieve the same ignition frequency.

Finally, it should be noted that for the Pittsburgh #8, the decreases in oxygen concentration shift the distributions to higher laser energies in approximately equal increments (equal energy ranges), and with little or no effect on the slope of the distributions. This finding is in contrast to the results for the Sewell coal (Fig. 3).

Three major differences between the ignition behaviors of the Pittsburgh #8 and Sewell exist. First, decreasing oxygen concentrations has a stronger effect in shifting the distributions of the Sewell to higher laser pulse energies (or higher particle temperatures). Second, as oxygen level is decreased, the slope of the distribution is undoubtedly decreased for the Sewell, while little effect is observed for the Pittsburgh #8. Finally, a comparison of the distributions of the two coals under 100% oxygen shows that the Sewell reaches 100% ignition frequency in a significantly smaller range of laser energy ( $\Delta E_{\text{laser}}$  of ~150 mJ versus ~250 mJ).

## DISCUSSION

Over the past three decades, many experiments have examined the ignition of pulverized coals under conditions which simulate pulverized fuel-firing conditions.<sup>4,5,6,7,8,9</sup> The common factor among these studies is the assumption of a single, average, kinetic rate-constant in describing the ignition reactivity of each coal. As we have shown previously,<sup>3</sup> it is necessary to account for the variation in reactivity among the particles within a sample in order to model the ignition distribution observed in this and nearly all previous ignition studies. Once such a model is implemented, the parameters may then be adjusted to fit the data and produce the desired ignition rate constant and reaction order with respect to oxygen for each coal.

Our previous experience in modeling ignition distribution data<sup>3</sup> provides some insight to explain the results described earlier. The model details will not be described here, but it is sufficient to note that the model accounts for particle-to-particle variations in reactivity by having a single preexponential factor and a Gaussian distribution of activation energies among the particles within a sample. The distribution is characterized by two parameters, an average activation energy ( $E_a$ ) and a standard deviation ( $\sigma$ ) in the activation energy.

In light of this model, the differences in the range of laser energies over which the various coals achieved 100% ignition frequency is a direct result of the breadth of the Gaussian distribution of activation energies: A narrow distribution (small standard deviation) leads to a small laser-energy range since most particles have similar activation energies and, thus, reactivities. Indeed, in the limit that the standard deviation is zero (all particles have the same activation energy), the ignition-frequency distribution would become a step function. Conversely, a broad distribution of reactivities (large  $\sigma$ ) leads to a relatively larger range of

laser energy needed to achieve 100% frequency, as is the case for the Pittsburgh #8 compared to the Sewell. The effect of variations in the average value of the activation energy in the distribution is to shift the ignition-frequency plot; higher  $E_{av}$  means lower ignition reactivity for a particular coal, which would shift the ignition distribution to higher laser energies.

Finally, with regard to the effect of oxygen concentration on the slope and shift of the ignition-frequency distributions observed for the Pittsburgh #8 and Sewell coals, the model interprets such differences to be the result of the variation in the reaction order,  $n$ , with respect to oxygen concentration.

The model results for the Pittsburgh #8 coal are shown in Figs. 4, 5 and 6, for average particle sizes of 115  $\mu\text{m}$ , 69  $\mu\text{m}$  and 165  $\mu\text{m}$ , respectively. The model parameters ( $E_{av}$ ,  $\sigma$ , and  $n$ ) were adjusted to produce the best fit at all oxygen concentrations for the 115  $\mu\text{m}$  particles (Fig. 4). They were then left at these values to produce the results for other particle sizes (Figs. 5 and 6).

As can be seen, while the model captures salient features in the experimental data, the fit to the data is not ideal. This is attributed to a lack of knowledge of the particle temperature at ignition. Indeed the results shown relied on calculated temperatures as described previously.<sup>1</sup> We plan to improve the experiment by implementing a two-color pyrometry system to directly measure the ignition temperature in future studies.

#### ACKNOWLEDGEMENT

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Coal Penn State ID	Rank	Prox. Analy. (dry wt%)		Ultimate Analysis (dry, ash-free wt%)				
		Vol. Matter	Ash	C	H	N	S	O (diff.)
Pittsburgh #8 (DECS 23)	low-volatile A bituminous	39.4	9.44	82.0	5.63	1.49	4.27	6.66
Sewell (DECS 13)	medium-volatile bituminous	25.0	4.22	88.2	4.95	1.50	0.65	4.71

Table 1: Ultimate and proximate analyses of coals used in this study.

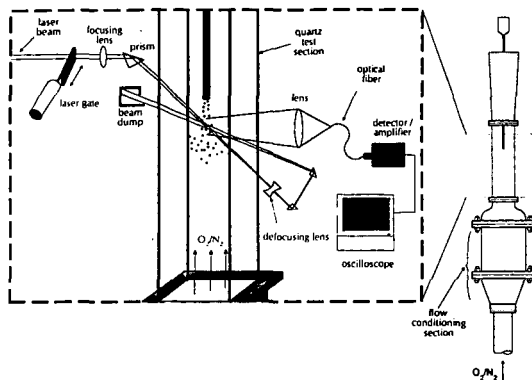


Fig. 1: Schematic of the laser ignition experiment.

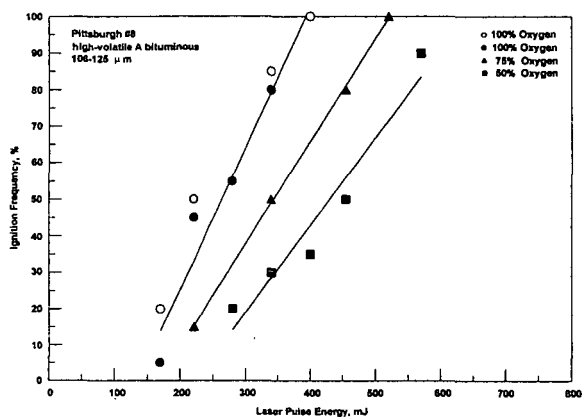


Fig. 2: Ignition-frequency distributions for the Pittsburgh #8 coal. Two data sets (open and filled circles) at 100% oxygen show reproducibility of experiment. Solid lines represent linear regressions of each data set.

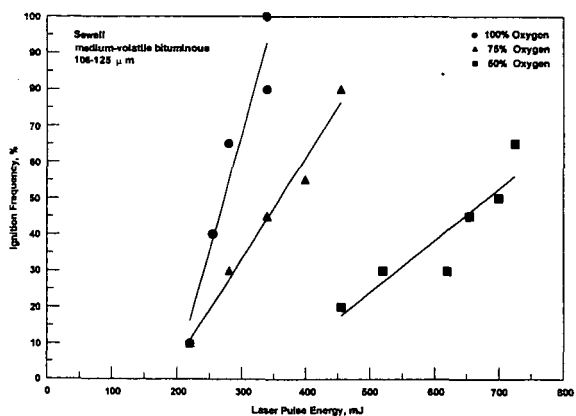


Fig. 3: Ignition-frequency distributions for the Sewell coal. Solid lines represent linear regressions of each data set.

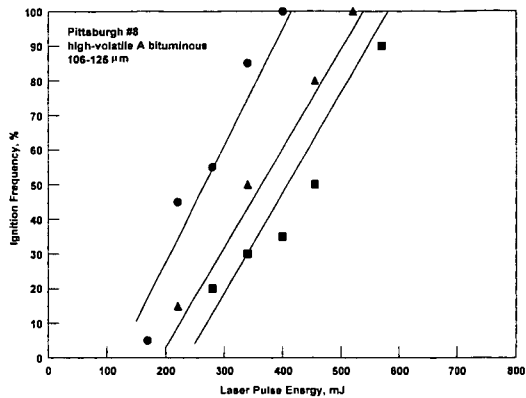


Fig. 4: Comparison of experimental data to model for 115  $\mu\text{m}$  particles. Lines represent linear regressions of experimental data and symbols are from simulation.

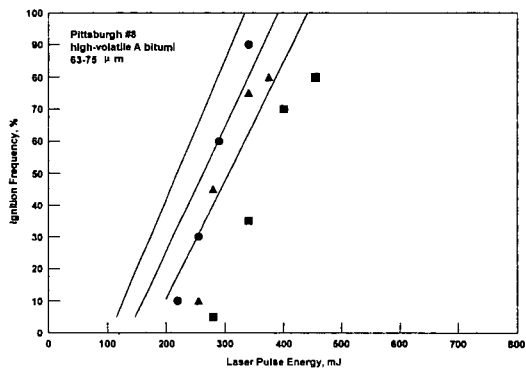


Fig. 5: Comparison of experimental data to model for 69  $\mu\text{m}$  particles. Lines represent linear regressions of experimental data and symbols are from simulation.

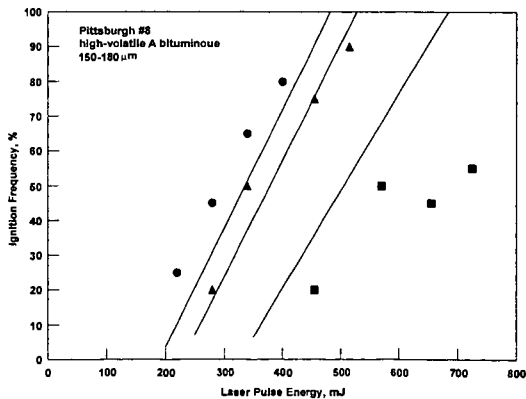


Fig. 6: Comparison of experimental data to model for 165  $\mu\text{m}$  particles. Lines represent linear regressions of experimental data and symbols are from simulation.